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# Optically detected magnetic resonance of sharp luminescence from Si/Si<sub>1-x</sub>Ge<sub>x</sub> superlattices

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The sharp emission from three Si/Si<sub>1-x</sub>Ge<sub>x</sub> superlattices has been analyzed experimentally using optically detected magnetic resonance techniques. Two types of response were observed in samples grown by molecular-beam epitaxy and rapid-thermal chemical vapor deposition. First, two samples exhibited spectra due to carrier heating in the underlying Si at the condition of cyclotron resonance. No spin resonance was observed, consistent with the previous assignment of the photoluminescence to an impurity-bound exciton. Second, one sample showed spin resonance from a weakly coupled electron and hole. Hence, the recombination mechanism is different from other samples. Various mechanisms are discussed.

## I. INTRODUCTION

The rapid progress in the growth of Si/Si<sub>1-x</sub>Ge<sub>x</sub> superlattices is demonstrated by the improvements in both the electronic and optical quality of the materials and devices. Focusing on the optical properties, only a few years ago there were no convincing reports of photoluminescence (PL) from these superlattices. However, since 1990 broad emission bands<sup>1-3</sup> and sharp emission bands<sup>4-6</sup> have been reported. In the past year, many groups have achieved sharp emission from these superlattices.

Since a simple PL spectrum does not reveal the underlying recombination process, we have applied optically detected magnetic resonance (ODMR) to learn more. ODMR probes the magnetic *g* factors of the recombining electron and hole, and directly measures the exchange interaction between the two particles. Previous ODMR has shown the band-edge character of the electron and hole producing the broad emission band,<sup>3</sup> and pointed toward donor-acceptor pair recombination as the underlying mechanism.<sup>7</sup> An alternate mechanism has been proposed based on platelets observed by transmission electron microscopy (TEM).<sup>8</sup>

The purpose of this article is to describe the application of ODMR to sharp emission bands from Si/Si<sub>1-x</sub>Ge<sub>x</sub> superlattices. Sharp PL containing a no-phonon line and TO and TA phonon replicas denotes emission that is band-edge related since these phonons connect the *k* vectors for the  $\Delta$ -point electron and the  $\Gamma$ -point hole. However, even for sharp emission a variety of mechanisms are possible and hence the ODMR can play a role.

In the following sections, results on three samples grown by two different techniques are presented. For two of the samples, the strongest response arose from cyclotron resonance (CR) of carriers in bulk Si regions. The CR illustrates that many of the carriers recombining in the superlattices are diffusing in from regions of bulk Si. In the third sample, spin resonance was detected with a small

exchange interaction between the electron and hole. The small exchange indicates that the recombination is either due to an unbound or fluctuation-bound exciton, to a type II (cross-interface) process or to donor-acceptor pair recombination.

## II. SAMPLES AND PL

The three samples studied had similar periodicities with Si layers from 40 to 45 Å and alloy layers from 15 to 20 Å (see Table I). The Ge mole fraction in the alloys layers ranged from 0.25 to 0.35. The first was grown by rapid-thermal chemical vapor deposition (RTCVD)<sup>9</sup> at a temperature of 700 °C for Si and 600 °C for the SiGe alloy. The two MBE samples were grown at 500 °C. Powder diffractometry showed that all three are good superlattices.

The photoluminescence spectra for these samples include a no-phonon (NP) line and phonon replicas which are characteristic of materials with the Si band structure (see Fig. 1). The RTCVD sample has a linewidth of about one-half that of the MBE samples. The two MBE samples have some emission at energies below the sharp emission. The deep emission is significantly stronger in the sample designated MBE-1 than in sample MBE-2. Detailed photoluminescence studies of RTCVD samples<sup>5</sup> and MBE-1<sup>10</sup> have been reported. The sharp bands of sample MBE-2 were found to increase with excitation power at a rate slightly faster than the square root. The position of the no-phonon line was found to increase with power at a rate of 0.75 meV/decade.

The energies of the NP lines have been compared to the expected energies for each sample. The nominal structure and composition values given in Table I were used. The band gap was taken from recent photoluminescence results<sup>11</sup> extended to  $x=0.35$  and the hole confinement energy was calculated in an envelope-function approximation. The energies for the MBE samples are higher than

TABLE I. Sample parameters.

| Sample          | Cap thickness (Å) | Si thickness (Å) | Si <sub>1-x</sub> Ge <sub>x</sub> thickness (Å) | x    | Number of periods | No-phonon energy (eV) | Excitonic band gap (eV) |
|-----------------|-------------------|------------------|---|------|-------------------|-----------------------|-------------------------|
| 821-3 (RTCVD)   | 150               | 45               | 20  | 0.35 | 30                | 0.955                 | 0.970                   |
| 00201.1 (MBE-1) | 300               | 40               | 20  | 0.25 | 83                | 1.068                 | 1.035                   |
| CH72 (MBE-2)    | None              | 45               | 15  | 0.35 | 100               | 1.047                 | 1.016                   |

expected. This discrepancy is attributed to Ge segregation distorting the profile of the alloy layers.<sup>12</sup>

### III. OPTICALLY DETECTED MAGNETIC RESONANCE EXPERIMENTS

#### A. Background

The samples were studied in an ODMR spectrometer based on a 7-T Oxford split-coil superconducting magnet. Faraday geometry was employed. Samples 2 mm in width were inserted into a 35-GHz TE<sub>011</sub> cylindrical microwave cavity at the position of maximum  $B_{rf}$ . However, because of their extent and high dielectric constant, the samples do sense a microwave  $E$  field. The PL was excited by different lines from an Ar-ion laser. The emission was separated either with a filter or a 0.25-m monochromator and detected with a cooled Ge detector. The microwave power

was on/off modulated and the change in emission coherent with this modulation was detected as a function of magnetic field.

Two types of response occur under these experimental conditions. The first is generally referred to as optically detected cyclotron resonance (ODCR).<sup>13</sup> It occurs when carriers (electrons or holes) are accelerated by the microwave electric field under the condition of cyclotron resonance, which is

$$\omega_c = eB/m^*, \quad (1)$$

where  $m^*$  stands for either the electron, light-hole, or heavy-hole effective mass. The magnitude of the effect depends on the mobility of the carrier. The photoluminescence is affected either through a change in the capture process or through impact ionization of an exciton. Although the linewidth does reflect the mobility of the electron or hole, it is not simply related to the true mobility. The second type of response is called optically detected magnetic resonance (ODMR).<sup>14</sup> It occurs due to  $B_{rf}$ -induced spin transitions between excited states with different recombination probabilities. Hence it requires a spin-dependent recombination. The condition for spin resonance between excited states is

$$\hbar\omega_s = g\mu_B B \pm 3c/4, \quad (2)$$

where  $g$  refers to either the  $|1/2, \pm 1/2\rangle$  electron or the  $|3/2, \pm 3/2\rangle$  hole and  $c$  is the exchange interaction between the particles.<sup>7</sup> The available microwave power limits the rate at which spin transitions can be induced. Thus, ODMR can only be detected for excited states with lifetimes long enough for spin-state equalization. In our spectrometer, ODMR can only be detected for lifetimes greater than 0.1  $\mu$ s.

Only the ODMR probes the recombination process directly. Hence, it is far more useful than ODCR for determining recombination processes.

#### B. Results for samples exhibiting ODCR

The response of sample MBE-1 in the ODMR spectrometer at different emission energies is dominated by cyclotron resonance (see Fig. 2). These data were taken with 458-nm excitation which penetrates well into the underlying Si. The Si emission (1.09 eV) is strongly decreased by cyclotron resonance. From the effective masses, the carri-

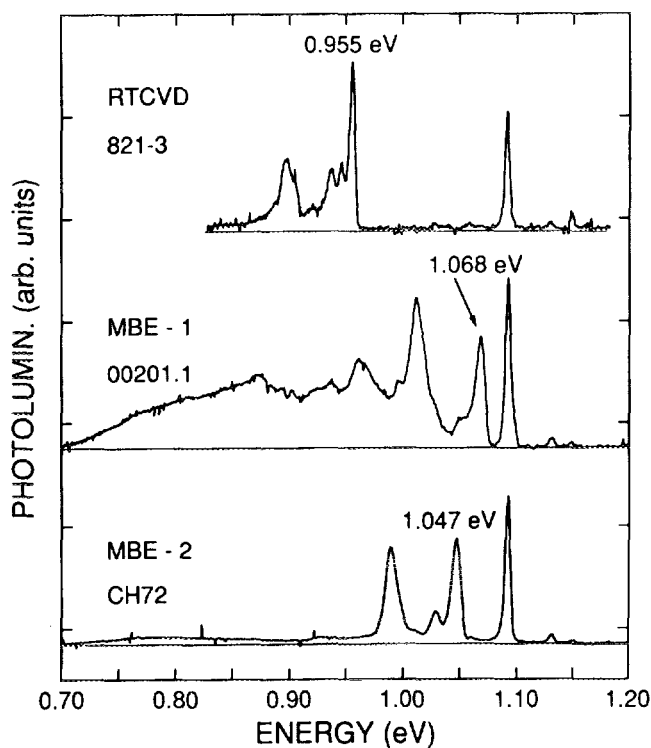


FIG. 1. PL spectra. The no-phonon line for the sharp emission from each superlattice is labeled by its energy.

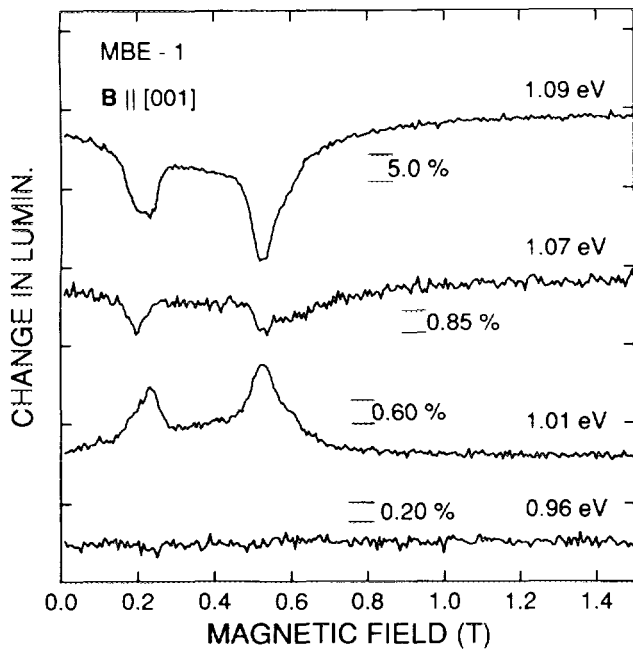


FIG. 2. Resonance spectra for MBE-1 with 458-nm excitation. The emission was analyzed by a spectrometer with a resolution of 27 meV at 1 eV. The Si emission at 1.09 eV is strongly decreased by the cyclotron resonance of electrons. The superlattice emission at 1.01 eV is enhanced by the same CR process. The deeper emission shows no detectable resonance.

ers are identified as electrons in the bulk Si regions. The NP line (1.07 eV) exhibits a small negative CR probably indicating a mixture of the higher and lower energy responses due to the low spectrometer resolution. The response at 1.01 eV is an increase in emission at CR, showing

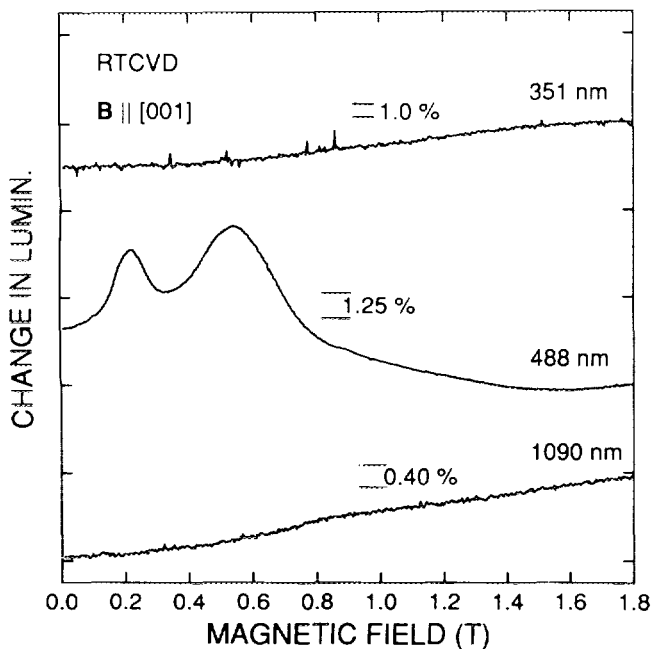


FIG. 3. Resonance spectra for the RTCVD sample for different wavelengths of excitation. With visible excitation, the emission for  $E < 1.0$  eV is enhanced by CR in bulk Si. Excitations which avoid the underlying Si produce no detectable resonance.

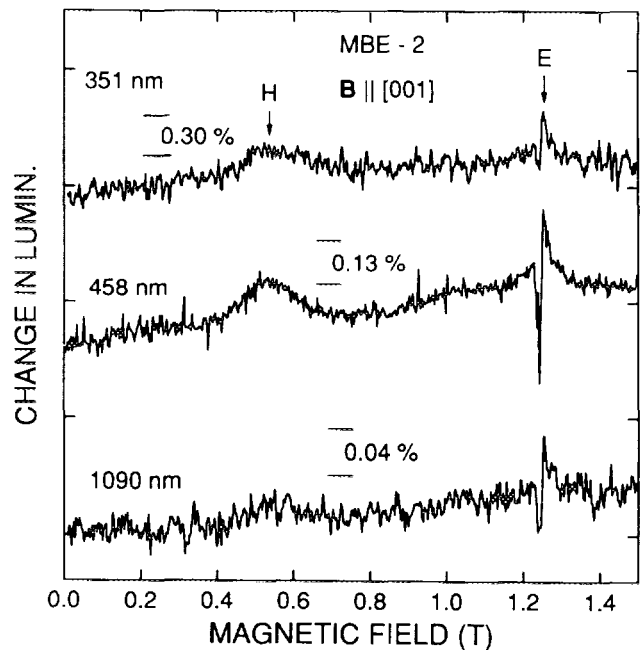


FIG. 4. ODMR (spin resonance) for MBE-2 for different wavelengths of excitation. The top spectrum was taken with a 1.3- $\mu$ m short-pass filter to exclude the broad emission. The middle spectrum was obtained with a 1.0- $\mu$ m long-pass filter. The lower spectrum was taken with the 0.25-m monochromator. The reduced signal strength for 1090-nm excitation probably indicates that some of the laser light passed through to the detector. Separate electron ( $E$ ) and hole ( $H$ ) resonances were detected. The negative signals near 1.25 T are due to point defects which compete with the radiative recombination.

that the SiGe emission benefits from the CR in the Si. This behavior indicates the diffusion of carriers from Si to the superlattice with recombination in the superlattice. At 0.96 eV, no CR or spin resonance is evident.

When visible light was used for excitation, sample RTCVD gave similar results to those for MBE-1. Hence different excitation wavelengths were used to enhance the sensitivity for spin resonance (see Fig. 3). For these experiments, a long-pass filter was used to exclude the emission from Si. Visible excitation still gave evidence of excitation in the Si regions with recombination in the superlattice through the positive CR signals characteristic of electrons and holes in Si. With UV excitation, the penetration depth is about equal to the Si cap thickness of this sample. Neither CR nor spin resonance was detected. The 1090-nm radiation is only absorbed by the alloy layers. Again neither CR nor spin resonance was detected.

### C. Results for the sample exhibiting spin resonance

Although sample MBE-2 exhibited CR effects similar to the other samples, the effects could be circumvented by taking advantage of its greater thickness. This sample also exhibited spin resonance which can be the dominant response under certain excitation and detection conditions (see Fig. 4). Positive (luminescence-enhancing) signals are observed clearly with UV excitation. A spectrum taken with visible excitation and detection of all emission below 1.2 eV is dominated by spin resonance through the cancel-

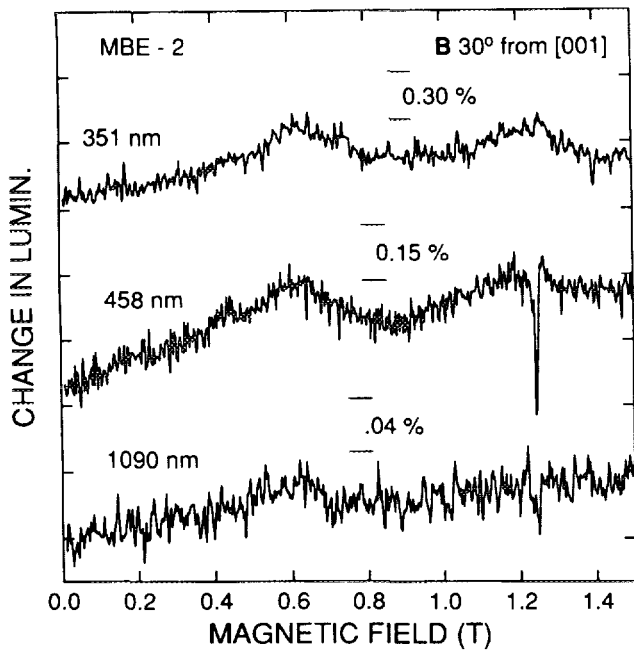


FIG. 5. ODMR for MBE-2 with the magnetic field rotated  $30^\circ$  from the superlattice axis. The conditions are the same as for Fig. 4. The hole resonance shifts to a higher field and the positive electron resonance is weaker.

lation of the negative CR on the Si emission with the positive CR on the superlattice emission. Finally, the resonance can be detected with IR excitation although the fractional response is smaller probably indicating that some of the laser line is getting through the spectrometer.

The line at 0.55 T shifts to higher fields as the magnetic field is rotated from the superlattice axis (see Fig. 5) and is assigned to the recombining hole. The  $g$  value for  $B$  parallel to the superlattice direction is  $4.7 \pm 0.1$ . The line shape of the hole resonance is different from that observed in wider-period samples whose emission spectrum is broad.<sup>7</sup>

Both positive and negative signals occur near 1.25 T which is the region of  $g=2$ . The negative signals are at least partially due to dangling bond defects which act to decrease the radiative emission.<sup>15</sup> The line shape of the positive part is distorted by these negative signals. Hence it is difficult to assign a  $g$  value or width to the positive-going part. Nonetheless, the existence of a positive-going signal near  $g=2$  is indicative of spin resonance from the recombining electron.

#### IV. DISCUSSION

The ODCR observed in each sample when excited by visible light provides information on the excitation of the superlattice. The penetration depth for visible light is approximately  $1 \mu\text{m}$ , which is greater than the thickness of each superlattice. The underlying Si has a high mobility and carrier heating by CR occurs readily. While the carrier heating decreases the probability of recombination in the Si region, it increases the chances that the carrier will diffuse

to the superlattice to recombine there. Hence, the CR is negative on the Si emission and positive on the superlattice emission.

Previous work has assigned the sharp emission in the RTCVD sample<sup>5</sup> and in MBE-1<sup>10</sup> to excitonic recombination at neutral shallow impurities. The lifetimes for this recombination in Si, which are determined by the nonradiative Auger process, are  $1 \mu\text{s}$  or less.<sup>16</sup> ODMR for processes this fast would be difficult to detect with our spectrometer. Hence, the lack of observed spin resonance is consistent with the previous assignment to an impurity-bound exciton.

The observation of spin resonance in sample MBE-2 points to a different recombination mechanism. The lifetime is likely to be longer than  $1 \mu\text{s}$  since signals of typical strength were observed. The broad linewidths of the electron and hole could arise either from inhomogeneous  $g$  value or from an exchange splitting between the electron and hole. Since separate electron and hole resonances are observed, the exchange interaction must be smaller than the linewidth. Using Eq. (2) for the hole resonance, this implies

$$c \leq (2/3)g_h\mu_B\Delta B. \quad (3)$$

With a linewidth of about 0.1 T and  $g_h=4.7$ , the exchange  $c$  is  $\leq 25 \mu\text{eV}$ .

There are three recombination processes with long lifetimes which can be considered for the emission from MBE-2. First, the free exciton in Si has a binding energy of 14 meV and might be expected to have an exchange coupling of about  $14 \mu\text{eV}$ .<sup>17</sup> A free exciton in  $\text{Si}_{1-x}\text{Ge}_x$  or an exciton bound to a fluctuation in composition, would have about the same exchange. Second, type II, or cross-interface recombination in AlAs/GaAs superlattices, exhibits exchange splittings from 1–34  $\mu\text{eV}$ .<sup>18</sup> Third, shallow-donor to deep-acceptor recombination in Si exhibits very small exchange coupling ( $< 0.05 \mu\text{eV}$ ) for a donor concentration of  $1 \times 10^{15} \text{ cm}^{-3}$ .<sup>19</sup> Each of these mechanisms is a candidate to explain the recombination in MBE-2 and further study is required to make the choice.

In summary, studies of samples with sharp emission using ODMR techniques reveal both ODCR and ODMR processes. The cyclotron resonance reveals the diffusion of carriers from the underlying Si to the superlattice. The absence of spin resonance in two samples is consistent with excitonic recombination at neutral impurities. The spin resonance in the third sample denotes a different process, but the particular process cannot be identified from the evidence presently available.

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